ON THE NATURE OF THE LATENT IMAGES FORMED IN PHOTOGRAPHIC EMULSIONS DUE TO LIGHT ABSORPTION AND TO THE PASSAGE OF IONISING PARTICLES*

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ABSTRACT. In this paper a comparison is made between the mechanism of latent image formation in silver halide grains present in photographic emulsions by photons and by ionising particles. It is shown that in the former the action of light proceeds from the surface of the grains inwards, while the energy transferred by ionising particles is a volume effect, starting the process of latent image formation from inside to the surface of silver halide grains. Thus the one favours the formation of surface and other of internal latent images. From this difference some of the observed differences in the action of photons and ionising particles can be explained, *e.g.*, emulsions with large grained particles will be more sensitive to photons than those with fine grained particles and *vice versa* for ionising particles. For the same transference of energy by photons and ionising particles to silver halide grains, the former will be more effective. The *rôle* of optical desensitisors in increasing the sensitiveness of fine grained emulsion to the action of ionising particles is also discussed.

An account is given of the theoretical deduction of H and D curve by Webb, which is based upon the assumption that the photographic emulsion contains silver halide grains with different degrees of photon sensitiveness.

Experimental evidence is discussed which shows that similar variation in sensitiveness to the action of ionising particles exists in silver halide grains in photographic emulsions. A theoretical H and D curve is deduced for the action of ionising particles and is compared with an experimental curve drawn for Ilford Halftone plates containing tracks of protons and α particles of different energies.

INTRODUCTION

It is well-known that a photographic plate consists of an emulsion of silver halide crystals in a film of gelatine. When such a plate is exposed to a light source of suitable wave-length, some of the silver halide crystals absorbing the light radiation become developable, *i.e.*, under the action of developers the affected silver halide grains are reduced to silver, which is deposited in the form of black grains. It is supposed that a latent image has been formed on the photographic plate which is acted upon by developing solutions. Large numbers of investigations have been directed towards elucidating the nature of the latent image formed due to light-absorption. Another subject which has been recently much discussed and which has an important bearing on our present review is the question of the sensitiveness of the photographic emulsion. If the different

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strips of a photographic film are exposed to a light source of constant intensity but the time of exposure is increased in a geometric progression, the plate, when developed, will show patches of different degrees of blackness or opacity. A curve is then drawn giving a relation between the density of deposit as function of the total quantity of light incident, It. Such a curve is known as H and D curve, which is a measure of the light sensitivity of the given film. Attempts have been made to interpret theoretically the dependence of the density of deposit in the exposed film on the integrated light intensity. Such interpretation can be made on two different assumptions: (*a*) the silver halide grains are all of the same sensitiveness, and (*b*) the different grains have different light sensitiveness, *i.e.*, they require the absorption of different numbers of photons to become developable.

Latent images are also formed in such films under the action of ionising particles. Investigations have been undertaken to find out how far the Reciprocity law holds good under such conditions. We are, however, more interested in studying the density of deposit of silver grains along the tracks of some of the ionising particles like α-particles, protons, and deuterons. It has been found that not all photographic emulsions are suitable for recording the tracks of ionising particles. Some like Ilford R_1 plates record only the tracks of α-particles, others like Ilford R_2 record, in addition, the tracks of protons and deuterons, while a third kind Ilford New Halftone plates records, in addition, tracks of mesotrons. It is further found that fast plates are not suited for this type of work, but on the other hand fine grained slow plates with large gradation like process plates and halftone plates are more suited. Further, it has been noted that the sensitiveness of plates like Imperial Process and Ilford R_2 to protons is considerably increased by bathing these plates in a dilute solution of pinakryptol yellow which is known to act as desensitisor of plates to the action of light. Another result has been obtained by the action of ionising particles on photographic emulsion which has a direct bearing on the question of the variability in the sensitiveness of silver halide grains in photographic emulsions. Thus, while it is found that the number of silver grains deposited per unit length of ionising tracks of α-particles is independent of the energy of the α-particle (Th C′ 8·95 MV, Po 5.40 MV), the number is found to be dependent on energy of protons and mesotrons. In the latter case it is found that within the certain energy-range the mean grain number is inversely proportional to the energy of proton particles. (Chowdhuri, 1941.) It is the purpose of the present paper to make a comparative study of the nature of the latent images formed under the action of light-absorption and of ionising particles. Further, the question will also be discussed in light of results obtained with photons and with ionising particles as to the variability in sensitiveness of silver halide grains.

The subject will be treated as follows :—

A—Latent image due to photon absorption :

§ 1—Mott and Gurney's theory ;

§2—Surface and internal latent images and chemical methods of distinguishing between them ;

§3—Distribution of sensitivity amongst silver halide grains ;

§4—Webb's theoretical deduction of H and D curves.

B—Latent image due to the action of ionising particles :

§5—Energy transfer to silver halide grains by ionising particles ;

§6—Evidence on the variability of sensitiveness amongst silver halide grains rendered developable by ionising particles ;

§7—Theoretical deduction of the H and D curve for ionising particles and comparison with experimental results.

A

§1. *Theory of latent image due to light absorption.*—When light is incident on silver halide grains, absorption starts from the outer boundary surface of the halide grain and proceeds inwards. According to the photochemical equivalence law, each photon absorbed will decompose one molecule of AgBr. The absorption takes place in the Br ion which leads to the release of the valence electron. According to the theory of Gurney and Mott (1938) this released electron will be raised to the conduction level of the AgBr crystal, where it is free to move about until it comes to the surface of the crystal and gets trapped on a so-called 'sensitive spot.' Each grain can have one or more sensitive spots which are supposed to be minute specks of silver sulphide, on which a trace of silver metal may have been deposited during the process of ripening. The concentration of a few such released electrons on a sensitive spot produces a strong local field in which some of the Ag^+ ions in the crystal grain move up and are deposited as silver atoms. This is the mechanism of the formation of latent image under light-absorption. The question as to the number of light quanta required to be absorbed before a latent image becomes developable will be discussed later.

When such an exposed plate is acted upon by a developer, the latter begins to act on the surface at the interface between the silver halide grain and the gelatine, the reduction spreading through the mass of the grain. It is supposed that the latent image acts as a heterogeneous catalyst which serves to distinguish between exposed and unexposed grains.

§2. So far we have considered only latent images formed on the interface of the silver halide grains. Recent investigations have shown that with increasing light intensity, latent images can also be formed inside silver halide grains. It is not possible by using normal developers to distinguish between surface and inner latent images. The technique which has been used by Berg, Marriage and Stevens (1941) for this purpose is as follows :-

(*a*) For surface latent image—Use a developer which contains as far as possible no alkali halide solvent, such as a glycine developer. With such a developer only the surface latent image is developed.

(*b*) For inner latent image—First use a bleacher to dissolve away only the surface latent image. This bleacher should not contain a silver halide solvent. Best results are obtained by using a solution of potassium bichromate in dilute sulphuric acid.

It is probable that a desensitisor like pinakryptol yellow acts as an oxidiser and bleaches the surface latent image.

An internal developer solution is made up, which contains a silver halide solvent in such concentration that the surface of the silver halide grain is etched away, exposing gradually the internal latent image which is then reduced by the developing solution.

With such a pair of developers, the relative concentration of surface and internal latent images as function of the intensity of exposure has been studied. It is found that the concentration of the internal to the surface latent image increases with the intensity of exposure.

Ordinary commercial developers usually contain sodium sulphite which is a silver halide solvent and with such developers it is not possible to distinguish between internal and external latent images.

§3. *Sensitivity of silver halide grains to light exposure.*—It is known from absolute sensitivity measurements that in an emulsion the most sensitive grains are rendered developable by the incidence of a few light quanta varying from one to twenty.

Recently a discussion has taken place in the Journal of Optical Society of America whether from a mathematical analysis of the H and D curve any information could be obtained as to the number of quanta required to form the latent image. Webb (1940) maintained that the conclusion arrived at must depend upon the basic assumption made in deriving the theoretical expression employed. It is shown how the assumption that all the grains require a fixed minimum number r of light quanta to acquire a latent image leads to the conclusion that only a small number of quanta is required for the formation of the latent image. It was further shown that if the alternative assumption is made that the grains in an emulsion have a wide range of sensitivity, the conclusion that a small number of quanta would be required for latent image formation would not follow. This statement led to a discussion between Webb and Silberstein which need not be considered here. To settle the question experimentally Webb and Evans (1941) performed an experiment in which H and D curves drawn for a film with strips exposed to intermittent light exposures It of different frequencies and intensities were compared to two similar curves due to (*a*) continuous exposures of intensity equal to that of the flash period of the intermittent exposure and (*b*) an exposure equal to the average intensity of the intermittent exposure. From a study of these curves the authors came to the conclusion that there were at least some grains in this emulsion which required between several hundreds to a thousand light quanta to produce developability.

§4. Having thus found an experimental verification of the assumption of the variability in light sensitivity of silver halide grains in a photographic emulsion, Webb (1941) proceeds to derive a theoretical expression for the variation of density of silver deposited in a photographic plate as function of exposure (H and D curve). We give a short account of his theory :

If during an exposure the number of light quanta incident per cm^2 on a photographic plate be E, then each silver halide grain receives on an average y units where $y = cE$: the maximum value of c is πa^2, the area of each grain. The number of photons absorbed by different grains will deviate from the average y, due to (*i*) inhomogeneity in size distribution of silver halide grains in the emulsion and (*ii*) fluctuation in the energy distribution in the incident light wave front. The probability that any grain receives n quanta, is given by Poisson's formula $w_n = \frac{e^{-y} \cdot y^n}{n!}$. Supposing now there is only one class of grains which require the absorption of at least r quanta to acquire developability, and if K is the total number of grains affected per cm^2 due to the given light exposure, then $\frac{K}{N} = \sum_{n=r}^{\infty} \frac{e^{-y} \cdot y^n}{n!}$; N is the total number of grains per cm^2.

If now there are different classes of grains requiring different number of light quanta, and the sensitivity distribution amongst them is given by a function $f(r)$ such that $\sum_{r_{min}}^{r_{max}} f(r) = 1$;

Then the fraction of the grains rendered developable due to an average exposure y is

$$\frac{K}{N} = \sum_{r_{min}}^{r_{max}} f(r) \sum_{n=r}^{\infty} \frac{e^{-y} \cdot y^n}{n!} . \qquad \ldots \quad (1)$$

The form of f (r) selected by Webb is $f(r) = a \cdot exp.\left[-k\left(ln \frac{r}{r_0}\right)^2\right]$

where
$$\frac{1}{a} = \sum_{r_{min}}^{r_{max}} exp.\left[-k\left(ln \frac{r}{r_0}\right)^2\right]$$

k is a parameter governing its overall latitude, and r_0 the maximum sensitivity.

For $\frac{K}{N}$ we can also write $\frac{D}{Dm}$, where D is the density $\left(\log_{10} \cdot \frac{1}{transmission}\right)$ for an exposure E.

Webb has compared his theoretical formula with the H and D curves of three different kinds of plates and has obtained the following values for the constants:

Plates	blue-sensitive	Average grain size, cm^2.	k	r_0	r_{min}
I.	Low Speed	0.115×10^{-8}	2.9	8	4
II.	Medium	0.300	.18	2.0	4
III.	High	0.700	.07	1/16	4

It will be noticed that for the same intensity of exposure E, the average number of quanta received per grain $y = cE$, is about six times for a grain in a high speed emulsion as compared to that for a low speed one. On the other hand it is found that overall gradation is much greater in the low speed plate; further, even in the high speed plates there are some grains which require the absorption of several thousands of light quanta to acquire developability. The H and D curve for the low speed plate is given in Fig. 1, Curve I, which is obtained on exposure to light of wave-length 4330 Å.

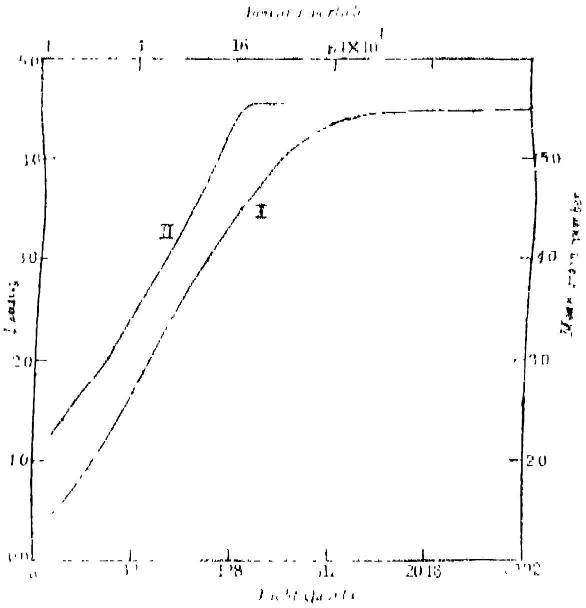

FIG. 1

The left hand vertical and bottom numbers refer to Curve I.

Abscissa—Mean number of light quanta absorbed per silver halide grain, λ = 4360 Å.

Ordinate—Mean density of developed plate.

The right hand vertical and top numbers refer to Curve II.

Abscissa—Mean energy absorbed per silver halide grain from ionising particle, in terms of light quanta units, λ = 4360 Å

Ordinate—Number of silver halide grains deposited per 10^{-3} cm. length of track of ionising particles.

§5. *Latent image formed by the action of ionising particles.*—When an ionising particle of mass substantially greater than that of an electron passes through a material medium, with not too great a velocity, so that the loss by radiation can be neglected, the energy lost by the particle will be chiefly due to collision with the bound electrons of the atoms in the material medium. The probability of transfer of energy by nuclear collision is small for ionising particles with small mass and small effective charge. The energy loss per unit length suffered by the particle is given by the following non-relativistic equation :

$$-\left(\frac{dT}{dx}\right)_{\text{coll.}} = 2\pi e^4 . z^2 . \frac{\sigma}{T} . \frac{M}{m} ln \left(\frac{4m}{M} . \frac{T}{I}\right) \qquad \ldots \quad (2)$$

T—kinetic energy of particle, whose mass is M.

ez—charge on particle.

σ—NZ = No. of electrons per unit volume of the medium.

I—13.6 Z = ionisation potential of the atoms.

If we express T in electron volts, and express M in terms of proton mass, then the energy loss of a particle of energy V electron-volts in a crystal of silver bromide is given by the formula

$$\text{Energy loss} = 6.01 . 10^2 z^2 . \frac{M}{V} \ln \left(1.04 \times 10^{-6} . \frac{V}{M} \right) \quad .. \quad (3)$$

The energy which is transferred in this way to the atoms of a silver halide grain will go to raise some of the bound electrons to the higher excitation levels including the conduction level of the AgBr crystal. Some of these will eventually be trapped on the sensitive spots present in the grain. Our knowledge about the mechanism of production of latent images in silver halide grains by ionising particles is not so detailed as of that formed by light-absorption.

Certain differences are however apparent. In the case of light-absorption, the process starts from the surface layer of the absorbing grain, and therefore the electron raised to the conduction level has a fair chance of being trapped on a sensitive spot which, it is assumed, is generally to be found on the surface. And the amount of energy absorbed is proportional to the surface area, *i.e.*, to a^2. For this reason also emulsions containing large-sized grains can more easily form latent images than those with small-sized grains.

In the case of the formation of latent images by the action of ionising particle, the ionisation produced is a volume rather than a surface one. If a is the radius of a silver halide grain considered to be spherical, then the average path of an ionising particle in such a grain is $p = \frac{4}{3} a$. The electrons which are set free by such a process have to traverse much longer paths before they can get trapped on the sensitive spots lying on the surface of the grains. The ionisation produced is proportional to the radius a, while the chance of meeting a sensitive spot on the surface is inversely proportional to a^2. This may be the explanation why slow Process and Halftone plates are more suited as detectors of ionising particles compared to fast plates containing large-sized silver halide grains.

It is worth investigating by the method described in §2 the relative ratio of surface to internal latent image formed in silver halide grains by the action of ionising particles. If it is found that the latter is much greater, then our interpretation of the relatively higher efficiency of emulsions containing small silver halide grains is valid. This would also explain the increase in sensitivity to the action of ionising particles produced by optical desensitisors like pinakryptol yellow, which by destroying the surface sensitive spot allows the internal latent images to come into prominence.

[Note added in proof—

H. Wambacher has recently published a report (Phot. Korr., **77**, 52, 1941) on the action of pinakryptol yellow (P) on photographic emulsion. From the very short summary given in Chem. Abs. (**36**, 5435, 1942) it appears that (*i*) emulsions treated with P before exposure give relatively small grains and (*ii*) with emulsion exposed to visible radiation and then treated with P, both the density and the number of developed grains diminish. These results are in agreement with our conclusions.]

The developers used for developing such plates like Elon-hydroquinone recommended by Wilkins (1940) contain sodium sulphite, which is a solvent for silver bromide. With such developers both internal and surface latent images are acted upon. It is proposed to undertake an experiment to determine the ratio of internal to the surface latent images produced by the action of ionising particles.

§5. We shall now consider in detail the developability produced in silver halide grains by the passage of ionising particles. In this case we have to consider a linear rather than a surface distribution of silver halide grains. We consider such particles to be spherical in shape and of radius of the order of 0.15×10^{-4} cm. All the particles whose centres lie within a cylinder of radius a are supposed to lie along the track of an ionizing particle whose trajectory is the axis of the cylinder.

The number of silver halide grains per unit length is not a constant, but is subject to a statistical fluctuation about its mean value s_0, given by Poisson's formula $w(s) = \frac{e^{-s_0}}{|s} \cdot s_0^s$. Further, if l is the length of the track of the ionizing particle through a grain, the energy transferred to it by collision is $-\frac{\partial T}{\partial x} \cdot l$; the value of l varies between o and $2a$. The energy transferred to each grain will fluctuate about the mean value $-\left(\frac{\partial T}{\partial x}\right)p$, where $p = 4/3.a$ is the average path of the ionising particles through the grains. The value of $\frac{\partial T}{\partial x}$ is given by formula (2). It is however not possible to make use of this formula, since the value of T is not known at each point on the particle track. We can however obtain indirectly an expression for the average energy transferred by the ionising particle to each silver halide grain on its path.

If T is the kinetic energy of the particle and L is its range in the emulsion, then the average energy loss per unit length is $\frac{T}{L}$. During the average path p of the ionising particle in each grain, the energy transferred is $\frac{T}{L} \cdot p \cdot \frac{\sigma_1}{\sigma_2}$, where σ_1 is the number of electrons per unit volume of a silver halide crystal and σ_2 is the number of electrons per unit volume of the photographic emulsion, *viz.*, $\sigma_2 = \Sigma n_k z_k$; where there are in the emulsion $1 \ldots k$ kinds of atoms of atomic numbers $z_1 \ldots z_k$ and their numbers present per unit volume are $n_1 \ldots n_k$ respectively.

It is found experimentally (Chowdhuri, 1941) that the number of grains deposited along tracks of α-particles of energy varying from 5.3 MV to 8.0 MV is an approximate constant independent of the particle energy. On the other hand the mean grain number along the tracks of protons for the energy range between 4MV to 10 MV varies inversely as the proton energy.

Thus we find that in spite of fluctuations in the number of silver halide grains along the tracks of ionising particles and the variation in the path traversed by the ionisation particles, certain mean values are obtained which indicate that the mean grain number deposited is inversely proportional, within a certain range, to the energy transferred to the silver halide grains

It is found that the energy received per grain increases with the diminution of the energy of ionising particle, and this is accompanied by an increase in the number of silver grains developed per unit length along the track. This indicates that the number of silver halide grains which become developable increases with the increase in energy received by them by collision with the ionising particle, *i.e., the silver halide grains present in the emulsion possess different degrees of sensitiveness.*

§7. We shall deduce an expression for the number of silver grains deposited along the track of ionising particles similar to that obtained by Webb for absorption of light quanta. We shall then give for comparison an empirical curve connecting the mean grain number deposited along the track of ionising particle as function of the mean energy absorbed by the silver halide grains

Let s be the number of halide grains per unit length in the emulsion, and s_0 the mean grain number; the value of s can be obtained by actually counting with a high magnification microscope along different directions on the plate If $f(r)$ is the fraction of the grains requiring r units of energy to acquire developability (measured either in electron volts or by the equivalent number of photons of a given wave-length), then the number k of grains rendered developable per unit length by the absorption of the mean energy r is given by $\frac{k}{s} = \int_{r_{min}}^{r} f(r)dr$; where r_{min} is the minimum amount of energy required to make a silver halide grain developable when struck by an ionising particle.

If for any kind of charged particle and for a particular range of energy the mean energy received by the silver halide grains r is greater than the amount r_{max} required to make the least sensitive of the grains developable, then within that range k the mean grain number deposited is independent of the energy of the ionising particle. This is the case we find for α-particles emitted by Po and ThC′ with energy-range from 5 to 9 MV. We shall, following Webb

assume that
$$f(r) = a\, e^{-k\left(ln\frac{r}{r_0}\right)^2}.$$

Then
$$\frac{k}{s} = a\int_{r_{min}}^{\bar{r}} e^{-k\left(ln\frac{r}{r_0}\right)^2}.\, dr \qquad \dots \quad (4)$$

is the theoretical form of the H and D curve for the formation of latent images due to ionising particels. *A priori* it can be said that the constants k, and r_0 for the same kind of emulsion will not be the same both for the action of photons and ionising particles. Further, as mentioned above, s is not a constant, but may vary for different tracks on the emulsion.

We shall now give an empirical H and D curve for ionising particles which is based upon the data given in Table I, part of which is taken from a paper by Miss Chowdhuri (1941), who has obtained a curve showing the relation between the mean grain spacing in Ilford Halftone plates of tracks of protons and α-particles of different energies.

TABLE I

Energy in 10^6 eV	Range in air in cm	Quanta absorbed per grain	Mean number of grains per 10^{-3} cm
Proton 1	2.3	8.36×10^4	1.5
2	7.1	6.06 ,,	4.0
3	14.1	4.00 ,,	3.57
4	24.0	3.20 ,,	3.43
5	34.0	2.83 ,,	3.03
6	47.0	2.45 ,,	2.90
7	61.0	2.21 ,,	2.78
8	77.2	1.99 ,,	2.70
9	95.5	1.82 ,,	2.60
10	115	1.07 ,,	2.50
particle 5.40	3.87	26.7 ,,	5.55
8.05	8.02	20.0 ,,	5.55

Column 1 gives the energies of proton and α-particles, whose ranges in R cm in air is given in Column 2, Column 3 gives the mean energy absorbed by silver halide grains expressed in terms of the equivalent number of light quanta of wave length 4300°A (energy 2.86 eV).

This is obtained from the formula,

$$r = \frac{T}{R} \cdot \frac{R}{L} \cdot p \cdot \frac{\sigma_1}{\sigma_2} \cdot \frac{\lambda e}{h.c} \tag{5}$$

where T—particle energy in electron volt,

R—range of particle in air,

$\frac{L}{R}$—ratio of ranges in the emulsion and in air $= 7.2 \times 10^{-4}$,

p = mean path of the ionising particle in the silver halide grains $= 0.2 \times 10^{-4}$ cm.

$\frac{\sigma_1}{\sigma_2}$ = the ratio of electrons present in unit volumes of silver bromide crystal, and of the emulsion $= 19.7$

$\frac{300hc}{\lambda e}$ = energy in electron-volt of photon of wave-length λ.

Column 4 gives the mean grain number per 10^{-3} cm of track in the photographic emulsion which is equal to $\frac{10}{m.g.s.}$ and is taken from Miss Chowdhuri's paper.

Fig. 1, Curve II is drawn from the data collected in Table I, and is of the shape to be expected from a sensitivity distribution curve of the form given by Webb. The experimental data are not sufficiently accurate nor sufficiently closed spaced in the region 4 to 30×10^4 quanta to enable us to determine the constants of the sensitivity distribution function. A comparison of the Curves I and II, drawn for sensitivity distribution curves of similar types of slow fine-grained plates as function of the number of light quanta and of collision energy absorbed, respectively, shows that light quanta are about 10 to 100 times more effective than ionising particles. This is what is to be expected considering the nature of the action of the two agents.

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